



Cumulative impacts of human activities on urban garden soils: Origin and accumulation of metals

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ABSTRACT

The concentration of heavy metals and soil properties in fifty urban garden soils of Szeged (SE Hungary) were determined to evaluate the cumulative impacts of urbanization and cultivation on these soils. Using two enrichment factors (EFs) (based on reference horizon; Ti as reference element) and multivariate statistical analysis (PCA), the origin of the studied elements was defined.

According to statistical coincidence of EFs confirmed by *t*-test, anthropogenic enrichment of Cu (EF = 4), Zn (EF = 2.7) and Pb (EF = 2.5) was significant in topsoils. Moreover, PCA also revealed the geogenic origin of Ni, Co, Cr and As and differentiated two groups of the anthropogenic metals [Pb, Zn] [Cu]. Spatial distribution of the metals visualized by GIS reflected the traffic origin of Pb; while based on ANOVA, the anthropogenic source of Cu is relevant (mainly pesticides) and there is a statistically significant difference in its concentration depending on land use.

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1. Introduction

Urban areas have expanded worldwide and are now occupying more and more lands which were formerly under agricultural use or natural conditions. The peculiarity of these modified soils is their special genesis, which occurs among special conditions not present in natural systems (Bullock and Gregory, 1991; Lehmann and Stahr, 2007; Norra and Stüben, 2003; Puskás et al., 2008). Therefore, urban soils have diversified physical, chemical and biological properties owing to various human activities (Rossiter, 2007).

Increase in the amount of heavy metals can be detected in the soils of the urban environment affected by a wide range of anthropogenic activities (e.g. disposal of municipal and industrial wastes, domestic heating, industrial emissions, heavy traffic and past land use etc.) (Kelly et al., 1996; Norra et al., 2001; Thornton, 1991; Wong et al., 2006). Heavy metals are of particular concern due to their long residence time in the soils and their toxicity to humans (Kabata-Pendias and Pendias, 2001).

The heavy metals in urban soils were first investigated in the late 1960s (Purves, 1967). The study of urban soil contamination has been in the focus of scientific research since then, giving impetus to world-wide study (Banat et al., 2005; Chen et al., 1997; Culbard et al., 1988; Imperato et al., 2003; Kelly et al., 1996; Lu et al., 2003; Madrid et al., 2002; Manta et al., 2002; Paterson et al.,

1996). Several investigations revealed that the metal contamination in cities is specific and varies with local conditions, but Zn and Pb are the most commonly enriched metals in the soils in urban areas.

Among urban soils, garden soils are recipients of further metal load from cultivation. Pesticides applied over a long period of time, compost, inorganic and organic manure, other soil improvers and contaminated irrigation water may result in metal accumulation (Csathó, 1994). Further possible sources of contamination in garden soils include: atmospheric deposition, paint particles, bonfires, contaminated material used for site levelling, runoff from metal surfaces, use of ash and mineral waste for constructing paths, burial of metal-containing wastes, leisure activities such as air gun shooting (Alloway, 2004).

Garden soils have a specific function: cultivating vegetables and fruits. Taken up by plants, heavy metals may enter the food chain in significant amounts. Therefore, people could be at risk of adverse health effects from consuming vegetables and fruits grown in soils containing elevated metal concentrations (Alexander et al., 2006). Additionally, heavy metals could possibly pose a risk to human health, when people ingest contaminated soil either on unwashed vegetables, on their hands, or, in the case of children, intentionally eating garden soil (Chaney et al., 1984).

Surveys of urban garden soils in several countries have shown wide ranges of concentrations of heavy metals, but these concentrations are in most cases higher than in natural or rural soils around the cities (Kahle, 2000; Moir and Thornton, 1989; Purves and Mackenzie, 1970; Wuzhong et al., 2004).

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Consequently, special attention should be paid to investigation of these soils owing to potential contamination and health damage.

The metal contents in the soils originate from natural (geogenic) sources based on the mineralogical and geochemical composition of parent material (lithogenic origin) and soil-forming processes (pedogenic origin) (Hindell and Fleige, 1989). However, beside natural origin, metals in the urban garden soils can derive from various anthropogenic sources. The mobility and plant availability of heavy metals depend on their origin, since plants do not readily take up lithogenic metals dominantly linked to primary minerals. By contrast, anthropogenic metals are mostly very mobile and readily accessible by plants (Kabata-Pendias, 1993).

Furthermore, metal mobility is also determined by several other factors, including some soil properties (pH, organic matter content and its quality, and clay content) (Szabó, 1996), which can also be strongly influenced by human activities.

Like many other major cities wide Europe, gardens are often located on the outskirts of Szeged (Mucsi et al., 2007), which serves as a good precedent from the viewpoint of above problems.

After considering the above facts, the major aims of the present study can be summed up as follows: (1) to determine the concentration of heavy metals and soil properties basically affecting metal mobility in urban garden soils of Szeged in order to evaluate the cumulative effects of urbanization and cultivation on these soils; (2) to distinguish anthropogenic metals from geogenic ones and define the degree of enrichment using enrichment factors (EF) and statistical methods; (3) to identify the origin of the anthropogenic elements by statistical analysis and spatial distribution of the metals.

2. Material and methods

2.1. Study area

The study area is located on the outskirts of Szeged (Baktó), situated in the inflow of River Maros into River Tisza. As the third largest city in Hungary, the population of Szeged is nearly 161 000. The climate is warm and dry with an annual mean temperature around 10.5 °C. The average annual radiation is between 2080 and 2090 h per annum with a mean annual precipitation of 520 mm (Marosi and Somogyi, 1990).

The natural soil types of the area have been transformed to different degrees: Technosols (FAO et al., 2006) are typical of the downtown, whereas human impact on the soils gradually diminishes from the downtown towards the peripherals (Puskás and Farsang, 2009). Consequently, Baktó in the north-eastern part of Szeged, covering nearly 1 km² area of family houses and private gardens, is situated in the transient zone between Technosols (FAO et al., 2006) in the downtown and natural soils around the city.

The plots in Baktó were distributed in the early 1930s; initially orchards were planted and due to urbanization and migration these were gradually transformed into a suburban residential area producing vegetables and fruits. Chernozems (FAO et al., 2006) were slightly modified as a result of the cultivation (intensive organic matter supplements, rotation, irrigation, etc.) and local anthropogenic activities (construction, infilling) (Szolnoki et al., 2011).

In addition to geogenic metals, anthropogenic metals contribute to the metal content of the soils in Baktó due to cultivation (metal-containing plant protection products, organic and inorganic fertilizers, soil improvers), atmospheric deposition and traffic emission from highway No. 47 between Szeged and Hódmezővásárhely (18 679 vehicle units/day) (Fig. 1).

2.2. Soil sampling

102 soil samples were taken from the gardens of 50 detached houses in 2010, with the prior authorization of the residents (Fig. 1). The studied gardens represented sites having different land use: vegetable gardens (31), orchards (9), and flower gardens (11). In the case of each garden, one composite sample (~1 kg) was taken from 10 to 12 topsoil samples (depth of 0–10 cm covering an area of 6–8 m²) using plastic instruments; one control point sample (depth of 80–100 cm) was collected from the middle of each plot with the help of a stainless steel manual auger. Moreover, a questionnaire on land use, historical data, applied pesticides and soil improvers and any infill in the given garden was filled in by the residents.

2.3. Soil analysis

Soil samples were air dried at room temperature and sieved to 2 mm then stored in closed plastic bags until analysis.

The pH (H₂O, KCl soil:solution ratio 1:2.5) was measured using a digital pH meter type Inolab pH 720. In order to capture the potential acidity of soils, the pH of a KCl soil suspension was also recorded. The carbonate content of dry soil samples given in percentage was determined via Scheibler type calcimetry. The total salt content was measured by recording the electric conductivity of fully saturated soil samples using a conductivity meter type OK-104. These methods were in accordance with the Hungarian Standard (MSZ), MSZ-08-0206-2 (1978). The texture was determined by the yarn test of Arany (MSZ-08-0205, 1978), which quantifies the amount of water in cm³ added to a 100 g air-dry soil sample to obtain a yarn (upper limit of plasticity) (Table 1).

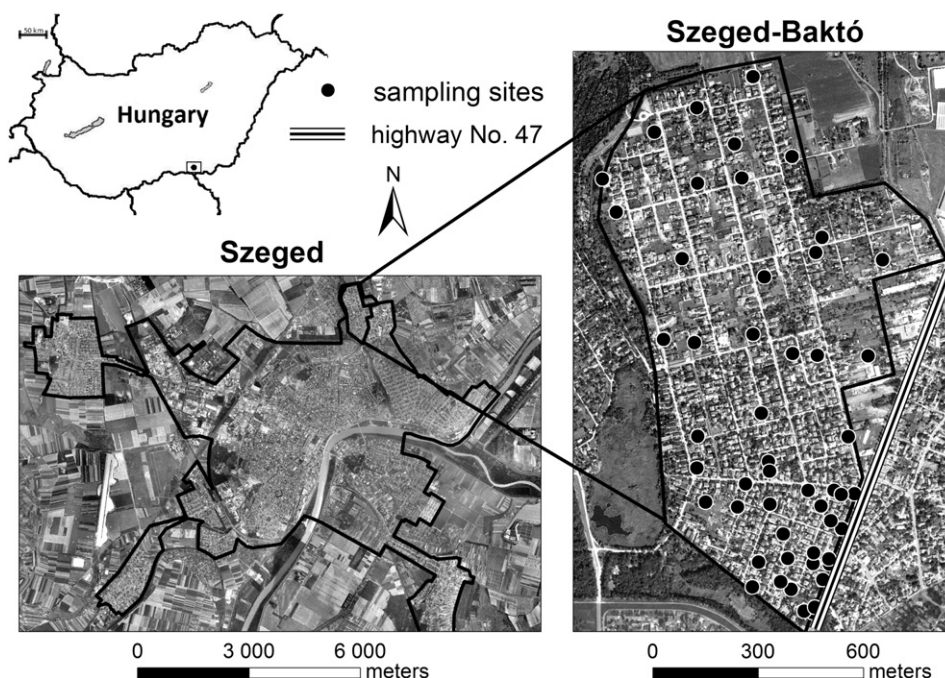


Fig. 1. Location of sampling sites in Szeged.

Table 1
Texture categories according to the yarn number of Arany.

Soil texture class	Yarn number of Arany
Coarse sand	<25
Sand	25–30
Sandy loam	30–38
Loam	38–42
Clayey loam	42–50
Clay	50–60
Heavy clay	>60

The organic content was measured after H_2SO_4 digestion in the presence of 0.33 M $K_2Cr_2O_7$ by spectrophotometer type Helios-γ (MSZ-21470-52, 1983). The humus quality of topsoils was given by the humus stability coefficient (K value) by Hargitai (Stefanovits et al., 1999). The basis of determining the quality of humus is that NaF and NaOH tend to dissolve differing organic compounds from the various humus components of soils. The raw and acidic humus fractions tend to dissolve primarily in NaOH solution, while the high-quality humus fractions of larger molecules enter into solution more easily in a NaF solution. The light-absorbing capacity of solution of soil samples treated separately with both chemicals was measured using photometry (E = extinction). When the comparison of the gained values (humus stability number) is divided by the total humus content (H) the result gives us the humus stability coefficient (K).

$$K = \frac{E_{NaF}}{E_{NaOH} \cdot H} \quad (1)$$

For the heavy metal contents, from each soil sample 0.5 g were digested with aqua-regia in closed vessels in a microwave oven (Anton Paar Multiwave 3000). Digestion in aqua-regia, which does not dissolve the silicate matrix, can give an estimate of the maximum amounts of elements that are potentially mobilized with changing environmental conditions. This method results in concentrations normally referred to as “pseudo-total” (Rao et al., 2008). Heavy metal contents (Cu, Ni, Pb, Cd, Co, Zn, Cr) and As together with Ti as a reference element were determined by inductively coupled plasma spectrometry (Perkin–Elmer ICP-OES Optima 7000 DV) using yttrium as internal standard (MSZ-21470-50, 2006).

2.4. Enrichment factors (EFs)

The EFs were calculated in order to assess the enrichment of the studied metals in the topsoil. Although there is no unique determination of the term “enrichment factor” in soil pollution studies, the EF approach has been widely used to evaluate the trace element enrichment in various soils (Blaser et al., 2000; Covelli and Fontolan, 1997; Facchinelli et al., 2001; Guerra et al., 2011; Massas et al., 2009; Sterckeman et al., 2006). In our study two types of EF were calculated.

The Top Enrichment Factor (TEF), according to Facchinelli et al. (2001), is the concentration ratio between the upper layer and the reference horizon. TEF was calculated with the following formula:

$$TEF = \frac{[E]_{SH}}{[E]_{RH}} \quad (2)$$

where $[E]$ is the concentration of the studied element (mg/kg), SH refers to the surface horizon (0–10 cm) and RH refers to the reference horizon (80–100 cm) in the same sampling point.

Table 2
Basic soil parameters of topsoils and control samples.

Sampling depth		Yarn number of Arany	Total salt content (%)	CaCO ₃ content (%)	pH (H ₂ O)	pH (KCl)	Humus content (%)	Hum. stab. coeff.
0–10 cm	N	51	51	51	51	51	51	51
	Mean	38.04	0.02	5.32	7.88	7.30	2.94	0.54
	Median	37.20	0.02	4.65	7.86	7.27	2.89	0.45
	Minimum	27.00	0.01	0.84	7.50	6.94	1.78	0.15
	Maximum	58.00	0.04	13.52	8.55	7.80	4.70	1.35
	Std. Deviation	5.39	0.01	2.52	0.25	0.17	0.75	0.32
	Skewness	1.16	1.09	0.97	0.84	0.56	0.63	1.05
	Kurtosis	2.92	1.75	1.09	0.41	0.55	−0.10	0.20
	N	51	51	51	51	51	51	—
80–100 cm	Mean	40.42	0.04	25.53	8.61	7.84	0.67	—
	Median	40.40	0.03	25.87	8.56	7.81	0.60	—
	Minimum	32.00	0.02	2.90	8.00	7.35	0.30	—
	Maximum	59.00	0.17	35.76	9.24	8.22	2.30	—
	Std. Deviation	4.00	0.03	5.69	0.31	0.17	0.36	—
	Skewness	1.59	3.55	−1.89	0.37	−0.13	3.49	—
	Kurtosis	8.64	14.12	5.55	−0.76	0.69	13.89	—
	N	51	51	51	51	51	51	—
	Mean	38.04	0.02	5.32	7.88	7.30	2.94	0.54

The Pedologic Enrichment Factor (EPF), according to Sterckeman et al. (2006), gives the proportion of the studied element and a suitably chosen reference element (normally Al, Li, Sc, Ti, and Zr) in the topsoil and in the parent material. The applied reference element was titanium (Ti), which is a common rock-forming element. Minerals of Ti are very resistant to weathering in the soil (Kabata-Pendias and Pendias, 2001). The pedologic enrichment factor was calculated as follows:

$$EPF_{Ti} = \frac{[E]_{SH}/[Ti]_{SH}}{[E]_{RH}/[Ti]_{RH}} \quad (3)$$

where $[E]$ is the concentration of any element, SH refers to the surface horizon (0–10 cm) and RH refers to the reference horizon (80–100 cm).

When EF values are around 1 or slightly below it, the elements have not enriched in the topsoil. Conversely, if this factor is greater than 1, there is metal enrichment in the topsoil due to soil-forming processes on the one hand and anthropogenic activities on the other hand. A natural pedogenetic enrichment is unlikely to produce EF values exceeding 2, while higher values point to an important anthropogenic input from the top (Facchinelli et al., 2001).

2.5. Statistical analysis and GIS

As it is difficult to evaluate the relative contributions of the metals from natural and anthropogenic processes based solely on enrichment factors, univariate and multivariate statistical analysis by PASW Statistic 18 software (SPSS Inc., 2009) were applied. Most of the multivariate statistical analysis and the statistical tests require normal distribution of data sets (Sajtos and Mitev, 2007), so logarithm transformation was carried out in the case of data with lognormal distribution. Graphical methods and a normality test (Kolmogorov–Smirnov test) were completed in order to assess the normality of original and transformed data (Reimann and Filzmoser, 2000). In multivariate analyses and applied statistical tests (t -tests, ANOVA), a database normalized by logarithmic transformation was realized.

Spatial analysis by GIS was also used to graphically and digitally present the distribution of the studied trace metals. A heavy metal distribution map and enrichment distribution maps were created using Arc Map 10 software.

3. Results

3.1. Evaluation of basic soil properties

The results of the questionnaires have revealed that 44% of the studied garden soils were filled or mixed with some soil materials, predominantly sandy material, in order to improve the structure of the original soil and elevate the surface level. However, both the degree and the kind of any infill were very heterogeneous; moreover their origin was usually unidentified. Kitchen and garden composts were also frequently mixed in the garden soils.

The texture of the studied soils was heterogeneous depending on the local infill using predominantly sandy material. Therefore, most topsoils were sandy loam or loam; some that originated from the northern part of study area could be characterized as clayey loam. By contrast, the control samples were mainly dominated by clay and clayey loam (Table 2).

Table 3

Metal concentrations digested with aqua-regia in the topsoils (0–10 cm).

	N	Mean (mg/kg)	Median (mg/kg)	Min. (mg/kg)	Max. (mg/kg)	Std. Deviation	Kurtosis	Skewness	"A" value (mg/kg)	"B" limit value (mg/kg)
As	51	7.19	6.65	3.06	15.89	2.33	2.90	1.16	10	15
Zn	51	80.17	74.00	32.82	198.71	30.86	3.81	1.57	100	200
Cd	51	0.55	0.48	0.27	2.86	0.36	33.30	5.39	0.5	1.0
Pb	51	15.71	13.78	5.11	60.85	8.23	17.68	3.48	25	100
Ni	51	22.62	22.48	10.04	35.60	4.70	0.92	0.19	25	40
Co	51	6.09	5.65	2.38	12.26	2.11	1.11	1.05	15	30
Cr	51	31.32	30.80	14.08	53.97	7.20	2.52	0.89	30	75
Cu	51	59.01	42.93	18.51	579.84	78.84	39.83	6.03	30	75
Ti	51	774.63	773.73	571.44	950.30	87.86	−0.29	−0.38	—	—

The average carbonate content of topsoils was 5.32%; the majority of samples could be considered moderately calcareous (2–10%), while the control samples (min. = 2.9%, max. = 35.8%) were highly (10–25%) and extremely calcareous (>25%) owing to loess parent material containing significant amount of lime, often in the form of concretion. The close correlation between the recorded pH values and those for the carbonate content was rather obvious. The pH (H₂O) values of topsoils (min. = 7.5, max. = 8.6) fell into the category slightly alkaline, whereas those of control samples could be classified into slightly alkaline and alkaline category (Stefanovits et al., 1999). A tendency for acidity is clearly discernible from the differences of the pH (H₂O) and pH (KCl) values. Difference 1 or around 1 between two pH values indicates easy vulnerability and higher tendency for acidity of the studied soils. The mean acidity value was 0.58 so the majority of studied soils have no inclined to acidity.

The humus content of topsoils (average = 2.94%, min. = 1.8%, max. = 4.7%) could be classified into diverse categories [poor (1–2%), moderate (2–4%), high (4–8%)] due to different cultivation techniques (compost, organic fertilizer, soil improvers of high organic content); whereas this parameter in the control samples (0.67%) belonged to the category extremely poor (<1%).

Besides the quantitative analysis, the humus quality (*K* value) was also determined in the topsoils. From the practical side, it is important to know the ratio of well-humified condense humus components composed of larger molecules, thus serve as metal bonding primary agents and have significant role in the environmental buffer capacity, to those organic components, which are not bond to calcium and less humified (Hargitai, 2008). The higher *K* value is, the better quantity the humus is. Compared to *K* value between 10 and 100 in high quality Chernozems (Stefanovits et al., 1999), the topsoils were characterized by the average *K* value of 0.54; the value exceeded 1 could be observed only in the case of seven gardens (max. = 1.35). All this indicated the prevalence of raw humus components, fulvic acids not yet subjected to humification. This fact could be resulted from infill on the one hand, the soil mixed with organic raw materials on the other hand.

The studied topsoils and control samples (min. = 0.01% max. = 0.17%) were slightly saline (0.05%–0.15%) (Stefanovits et al., 1999). Based on the above-mentioned, various soil properties, but the total salt, could be noticed in the study area (Table 2).

3.2. Heavy metal contents in the garden soils

In Hungary, Joint Decrees (6/2009. (IV. 14) KvVM-EüM-FVM and 10/2000. (VI. 2) KöM-EüM-FVM-KHVM) were created to protect the quality of groundwater and soils. Various limit values are defined by these decrees. Background concentration (A): Representative value, typical concentration of a particular substance reflecting natural, or close to natural, conditions in the soil. Pollution limit

value (B): Risk substance, with due regard to the case of the geological medium of the full range of soil functions and the sensitivity of groundwater to pollution. The concentrations of the studied components are compared with the above-defined "A" and "B" values (Table 3).

All the studied metals in the topsoils, except for Co, exceeded the "A" background concentration in the following proportion of the samples in per cent: Cu (82%), Cr (62%), Cd (38%), Ni (26%), Zn, Pb, As (<25%) (Fig. 2). Arsenic, Cd and Cu exceeded the "B" limit value in 2%, 4%, and 14% of the samples, respectively. Consequently, nine garden soils could be regarded as polluted.

3.3. Effect of the different garden types on metal concentrations

One-way analysis of variance (ANOVA) was applied to determine whether land use exerts any influence on metal concentration in the garden soils. In our work, the independent variable was the garden type (orchard, vegetable and flower garden), while the dependent variables were the recorded metal concentrations. As a result, similar concentrations were measured in the orchards, vegetable and flower gardens. Therefore, the facts clearly demonstrated that the garden type had no effect on the metal concentrations in the case of most metals (Table 4).

However, there was a statistically significant difference in the Cu concentrations of different garden types. Cu was significantly higher in the soils of orchards and vegetable gardens than in flower gardens ($p < 0.05$), whilst there was no significant difference in the Cu content between the soils in orchards and vegetable gardens (Fig. 3). This fact evidently confirmed that human activities (mainly copper pesticides), beside natural processes, could also play an important role in the Cu concentration of the studied soils.

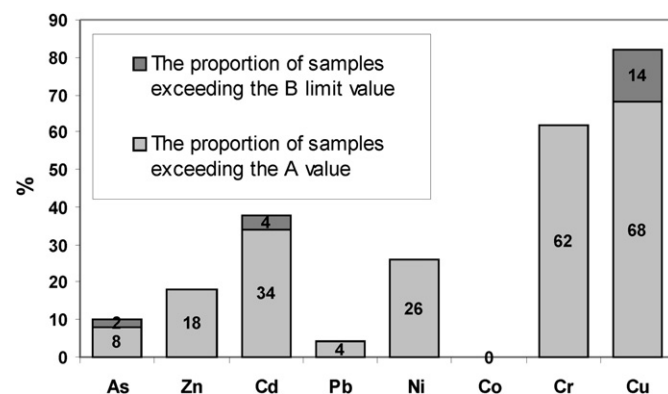
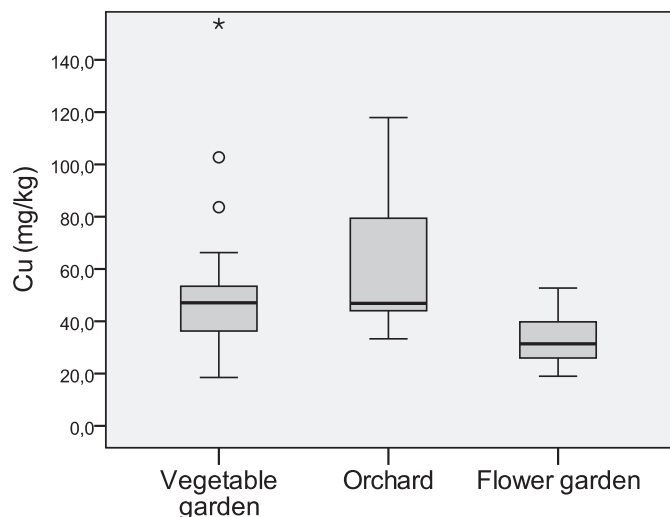


Fig. 2. The proportion of samples (%) exceeding the "A" background concentration and the "B" limit value of the studied elements.

Table 4

The average metal concentration (mg/kg) measured in the topsoil (0–10 cm) in different garden types.

Type of garden		As	Zn	Cd	Pb	Ni	Co	Cr	Cu
Vegetable garden N = 31	Mean (mg/kg)	6.86	75.70	0.47	14.46	21.97	6.23	29.85	66.62
	S.D.	2.47	22.50	0.11	5.32	4.90	2.54	7.17	98.68
Orchard N = 9	Mean (mg/kg)	8.30	87.63	0.78	19.24	23.31	5.83	34.04	64.54
	S.D.	2.34	38.80	0.80	16.12	4.68	1.60	9.60	31.99
Flower garden N = 11	Mean (mg/kg)	7.21	86.65	0.59	16.31	23.89	5.92	33.22	33.04
	S.D.	1.70	43.36	0.24	5.44	4.20	0.82	3.70	9.98

**Fig. 3.** Cu concentrations (mg/kg) measured in the topsoils (0–10 cm) in different garden types.

3.4. The impact of traffic emission on metal contents

An independent sample *t*-test, as a statistical procedure to compare the averages of two independent groups, and spatial distribution of the metals, were carried out in order to detect the metal contamination originating from a busy road (highway No. 47) situated near the study area. The studied garden soils were classified into two groups based on the distance (<10 m; >10 m) from highway No. 47. The averages of the above groups were compared afterwards in order to verify the widely known pre-hypothesis that heavy metals show elevated levels in the topsoil in the immediate vicinity of roads with relatively large traffic densities, but that the decrease in concentration with distance is rapid (Szegedi, 1999).

The averages of each metal concentration, except for Cu, were higher in the garden soils close to the busy road than in those farther from the road (Table 5). However, a significant difference ($p < 0.05$) could be noticed exclusively in the case of Pb.

The spatial distribution map on Pb depicted in Fig. 4 confirmed the above. For other metals, no clear relationship between distance from the road and spatial distribution could be observed.

Table 5

The average metal concentrations (mg/kg) in the topsoil (0–10 cm) near and farther from the road (the extreme values from every group were excluded).

Distance from road No. 47		As	Zn	Cd	Pb	Ni	Co	Cr	Cu
Distance from road <10 m	Mean (mg/kg)	8.25	79.30	0.55	23.15	23.75	6.75	33.50	42.99
	N	6	6	7	7	7	7	7	6
	Std. Deviation	1.42	15.51	0.06	4.35	2.67	1.28	3.82	7.77
Distance from road >10 m	Mean (mg/kg)	6.85	77.59	0.48	13.44	22.44	5.99	29.89	45.24
	N	44	44	42	43	44	44	42	42
	Std. Deviation	2.01	27.31	0.12	3.88	4.95	2.21	5.82	20.55

3.5. Metal enrichment in garden soils

$TEF_{average}(As)$, $TEF_{average}(Ni)$, $TEF_{average}(Co)$, $TEF_{average}(Cr)$ were around 1, indicating that these metals have not become enriched in the topsoil. $TEF_{max}(Ni)$, $TEF_{max}(Co)$, $TEF_{max}(Cr)$ were considerably below 2. $TEF_{max}(As)$ approached 2 in a garden soil where the As concentration exceeded the “B” limit value (Table 6). $TEF_{average}(Cd)$ was 1.43, reflecting moderate enrichment; in the case of some garden soils, $TEF(Cd)$ ranged between 2 and 3. However, $TEF_{max}(Cd)$ approached 5 in a garden soil where the Cd concentration exceeded the “B” limit value. $TEF_{average}(Pb)$ and $TEF_{average}(Zn)$ were 2.5, while $TEF_{average}(Cu)$ was 4, indicating significant enrichment of these metals in the topsoil (Table 6).

$EFP_{average}(As)$, $EFP_{average}(Ni)$, $EFP_{average}(Co)$, $EFP_{average}(Cr)$ were around 1, reflecting that these metals have not become enriched in the topsoil. $EFP_{average}(Cd)$ was 1.5, while $EFP_{average}(Pb)$ and $EFP_{average}(Zn)$ were 2.5 and $EFP_{average}(Cu)$ was 4, denoting significant enrichment in the topsoil (Table 7). However, $EFP_{max}(Cu)$ was around 30 in a garden soil exceeding the “B” limit value of Cu.

The linear correlation coefficient was 0.991 ($p < 0.01$), so the very strong relationship between the EFP and the TEF was unambiguous (Fig. 5). In the case of each metal, a paired samples *t*-test was applied in order to examine the coincidence of TEF and EFP values. There was no significant difference ($p > 0.05$) between EFP and TEF values; the average pairwise differences for all the metals were lower than 0.052. Consequently, both enrichment factors can be used for detecting the metal enrichment in the topsoil.

3.6. Statistical evaluation of the data

Principal component analysis (PCA) was performed for the dataset of topsoils so as to explore the relationship between the eight metals as variables, and to assign related variables into principal components.

On preliminary examinations, together with the multi-correlation, the significant Bartlett test ($p < 0.05$), and the criterion of Kaiser Meyer-Olkin ($KMO = 0.683$), our data seemed to be eligible for PCA. In the selection of factor numbers, the eigenvalue size and the variances explained were all taken into account. The factors were rotated using the varimax rotation to define the factor loading matrix more easily.

In the analysis, three principal components were considered, which account for over 75% of the total variance (Table 8). The

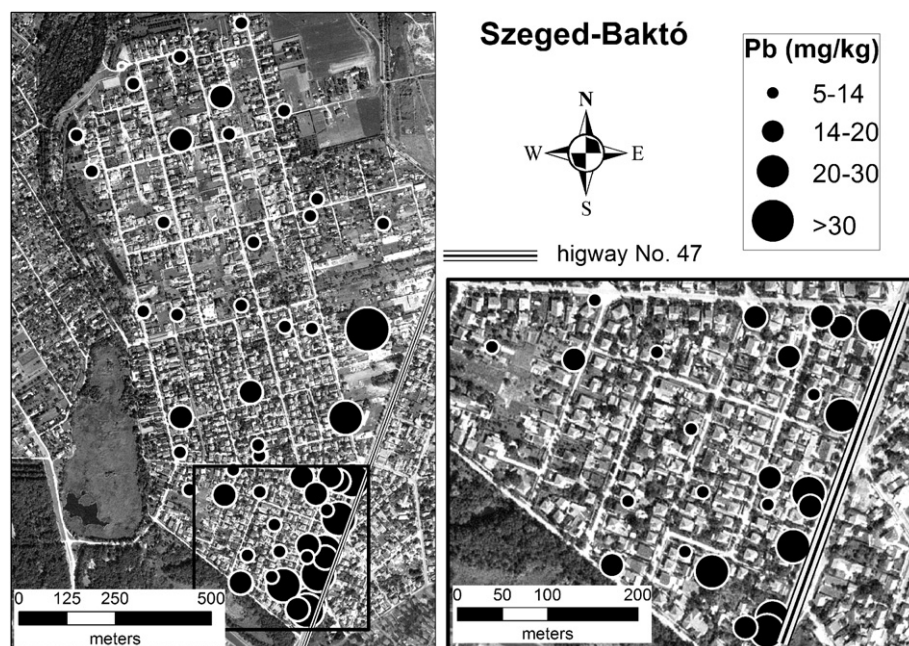


Fig. 4. Spatial distribution map on Pb.

eigenvalues of the first two extracted factors were higher than 1, and the third became greater than 1 after the factor rotation (Table 8). Pb, Zn, and Cd could be categorized into the first principal component (PC1), due to the high component loadings of PC1 in the rotated component matrix. The second and third principal components were composed of Ni, Co, Cr, As as well as Cu alone, respectively (Table 9).

4. Discussion

4.1. Metal contamination in urban gardens

Concentrations of As, Cd, and Cu, of the recorded metals, exceeded the “B” limit value in only 18% of all the studied garden soils. Consequently, the metal contamination in the study area can be considered to be moderate. Concentrations of As, Cd slightly, and Cu concentration significantly exceeded the “B” limit value; in the case of one garden, for example, Cu concentration is eight times higher than the limit value. Concentrations of Cu, Cd, and As exceeded the limit value exclusively in the orchards and the vegetable gardens; the flower gardens and the orchards; and the vegetable gardens, respectively.

Anthropogenic origin is unambiguous in the case of metals (e.g. Cu) exceeding the “B” limit value. Other metals that have not reached the “B” limit value but exceeded the “A” background

concentration may also originate from an anthropogenic source. However, the “A” background concentration can be used for only limited assessment of the anthropogenic enrichment, since it can be a lower or higher value depending on the geological background (Kádár, 2007). Cr, for example, supports the above facts well. Cr concentration exceeded the “A” background concentration in many topsoils, indicating a potential anthropogenic origin. However, the anthropogenic origin of Cr is non-evident due to metal values exceeding the “A” value in the control samples, too. Consequently, the relatively high amount of Cr in the topsoils is of geogenic rather than anthropogenic origin.

4.2. Anthropogenic metal enrichment in urban garden soils

We used the results of EFPs to visualize the spatial distribution of metal enrichments owing to the coincidence of TEFs and EFPs. The map of the spatial distribution of EFPs of the recorded metals in the topsoils depicted in Fig. 6 sharply differentiates between metals of anthropogenic and geogenic origin. In accordance with the EFP results, of all the studied metals, Cu was enriched the most significantly in the topsoils ($EFP_{average} = 4$; 92% of EFPs are higher than 2), denoting the anthropogenic origin of this element. Intense Cu accumulation can be observed all over the study area, but the highest values were measured in the soils of orchards and vegetable gardens situated in the middle of Baktó (Fig. 6).

Table 6

Top Enrichment Factor (TEF) calculated using reference horizon.

TEF	N	Mean	Median	Min.	Max.	Std. Deviation	Kurtosis	Skewness
As	51	0.89	0.84	0.38	1.80	0.32	0.21	0.76
Zn	51	2.72	2.57	1.12	7.34	1.07	5.80	1.70
Cd	51	1.43	1.31	0.78	4.79	0.59	21.72	4.05
Pb	51	2.54	2.33	0.52	5.34	0.97	0.39	0.33
Ni	51	0.95	0.94	0.40	1.51	0.20	1.49	0.34
Co	51	0.94	0.97	0.40	1.40	0.20	0.72	−0.42
Cr	51	1.07	1.06	0.47	1.71	0.21	2.48	0.61
Cu	51	4.16	3.32	0.79	29.03	4.04	29.64	4.98

Table 7

Pedogenic Enrichment Factor (EFP) calculated using Ti as reference element.

EFP _{Ti}	N	Mean	Median	Min.	Max.	Std. Deviation	Kurtosis	Skewness
As	51	0.90	0.80	0.36	1.85	0.36	−0.30	0.59
Zn	51	2.73	2.66	1.07	6.63	1.05	2.68	1.18
Cd	51	1.45	1.35	0.63	4.81	0.66	14.29	3.36
Pb	51	2.54	2.51	0.56	5.37	0.98	0.58	0.43
Ni	51	0.96	0.95	0.52	1.70	0.22	1.57	0.61
Co	51	0.95	0.96	0.39	1.39	0.21	0.21	−0.49
Cr	51	1.07	1.04	0.60	1.96	0.24	3.82	1.37
Cu	51	4.18	3.34	0.83	29.92	4.14	30.89	5.10

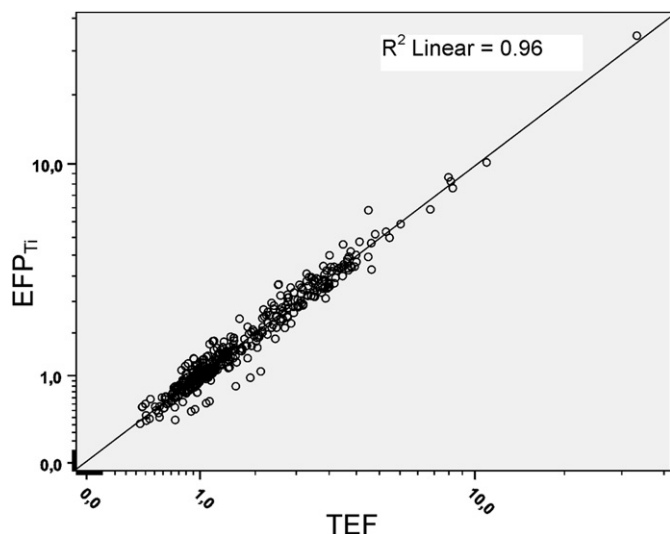


Fig. 5. Relationship between TEF and EFP.

In addition to Cu, Zn and Pb were also enriched in the topsoils ($EFP_{average} = 2.5$), indicating their anthropogenic origin. In spite of significant Zn and Pb enrichment in the soils close to the road, these metals are evenly distributed throughout the study area. Cd was slightly enriched in the topsoils ($EFP_{average} = 1.5$), but some higher $EFP(Cd)$ values (>2) reflect its anthropogenic origin. Therefore, anthropogenic enrichment of Cd may derive from point sources and heavy traffic (Fig. 6). Ni, Cr and Co were not enriched in the topsoils ($2 > EFP_{average} \sim 1$). Consequently, these metals can be considered to be of geogenic origin. Although there was one garden where As content exceeded the “B” limit value, As can also be regarded as a geogenic metal (Fig. 6).

During the principal component analysis, three principal components were identified to describe metal behaviour in our garden soils, confirming EF results (Fig. 7).

The metals with significant enrichment in the topsoils (Pb, Zn, Cd) fell into the first principal component (PC1), reflecting their anthropogenic origin.

The second principal component (PC2) comprises metals without enrichment ($EF \sim 1$) in the topsoils (Ni, Co, Cr, As). Anthropogenic origin of these metals can be excluded, and their concentrations in the topsoil are determined by the geochemical composition of the parent material and geogenic processes. Thus, the PC2 can be defined as the group of “geogenic metals”.

Cu, exceeding the “B” limit value in many cases and having the highest EF values regarding the studied metals, can be categorized into the third principal component (PC3). Consequently, it is obvious that Cu concentration is controlled predominantly by anthropogenic activities.

Table 9

Rotated component loading matrix.

Element	Rotated component matrix		
	PC1	PC2	PC3
Pb	0.874	0.216	0.120
Zn	0.838	0.086	0.187
Cd	0.638	0.406	−0.022
Ni	0.289	0.896	0.055
Co	0.034	0.736	0.287
Cr	0.499	0.698	−0.016
As	0.483	0.501	0.451
Cu	0.116	0.125	0.945

Bold numbers indicate the most important components of each principal component (PC).

Zn, Cd, Pb of the PC1 can come from organic fertilizers (Wuzhong et al., 2004); organic and inorganic fertilizers (Csathó, 1994); and some pesticides (Alloway, 2005), respectively. However, the major emission source of these elements is likely to be traffic emission. Higher Pb concentrations can be detected in the samples near the road, confirming the traffic origin of Pb. Nevertheless, the $EF(Pb)$ values verify that Pb accumulation is not limited to garden soils close to the road (Fig. 6). The relatively strong positive correlation ($r = 0.691$) between the Pb and Zn concentrations demonstrates that the excess Zn may also originate from traffic. This agrees with results obtained by several authors (Bretzel and Calderisi, 2006; Manta et al., 2002) who also found strong positive correlation between the Pb and Zn concentrations in urban soils (mainly in roadside soils), suggesting the dominant role of vehicle traffic as common anthropogenic source of these elements. However, in our sampling area the origin of excess Zn is very complex: in addition to the traffic emission and atmospheric deposition, the soil improvers and organic fertilizers can also be an important anthropogenic source of Zn. Cd is slightly enriched in the topsoil as traffic emits relatively small amounts into the environment. The major sources of Pb emission have historically been from fuels in on-road motor vehicles; most Zn input into the soils may be tyre wear, whereas Cd can be released to the atmosphere through tyre wear and fuel combustion (Hjortenkrans et al., 2006).

Cu, as the single element of PC3, has a significant moderately strong correlation with more metals in the control sample (data not shown), but no correlation with other metals in the topsoils, indicating specific sources of this element. Cu emission has significantly increased in recent years due to asbestos-free brake linings of road vehicles (Hjortenkrans et al., 2006; Salma and Maenhaut, 2006). Despite this fact, the most intensive Cu accumulation can be identified in the topsoils in the middle of the study area rather than of those near busy roads (Fig. 6). The potential anthropogenic sources of Cu are pesticides full of copper whose long-term use may result in an increase in the Cu concentration in topsoil. According to the *t*-test, higher Cu

Table 8

Total variance explained.

Component	Initial eigenvalues			Extraction sums of squared loadings			Rotation sums of squared loadings		
	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %	Total	% of variance	Cumulative %
1	4.059	50.736	50.736	4.059	50.736	50.736	2.454	30.669	30.669
2	1.015	12.685	63.421	1.015	12.685	63.421	2.317	28.965	59.634
3	0.930	11.621	75.042	0.930	11.621	75.042	1.233	15.408	75.042
4	0.701	8.762	83.804						
5	0.569	7.110	90.914						
6	0.378	4.721	95.635						
7	0.266	3.321	98.956						
8	0.084	1.044	100.000						

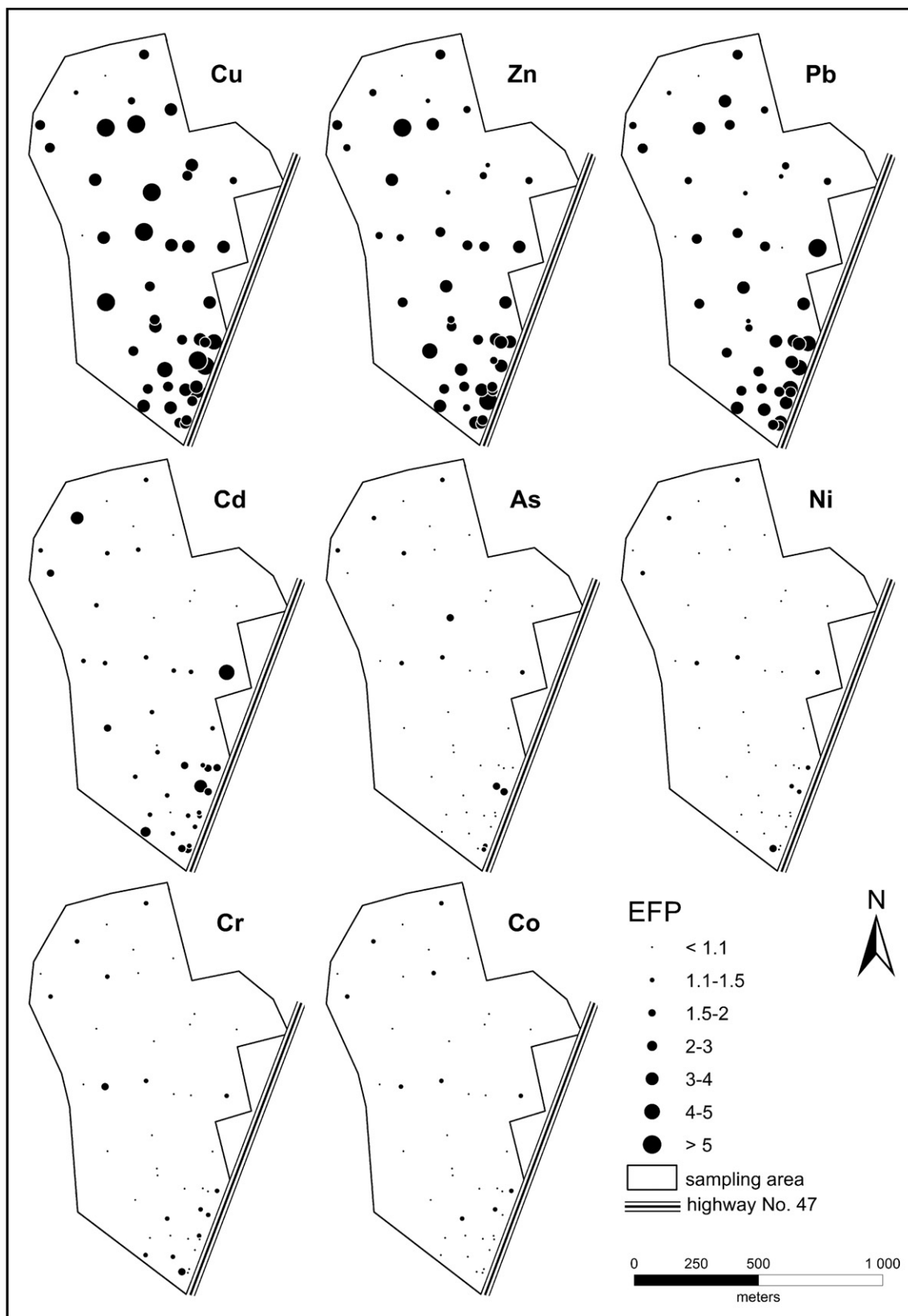


Fig. 6. The spatial distribution of pedological enrichment factor (EFP_{Ti}) calculated using Ti as reference element.

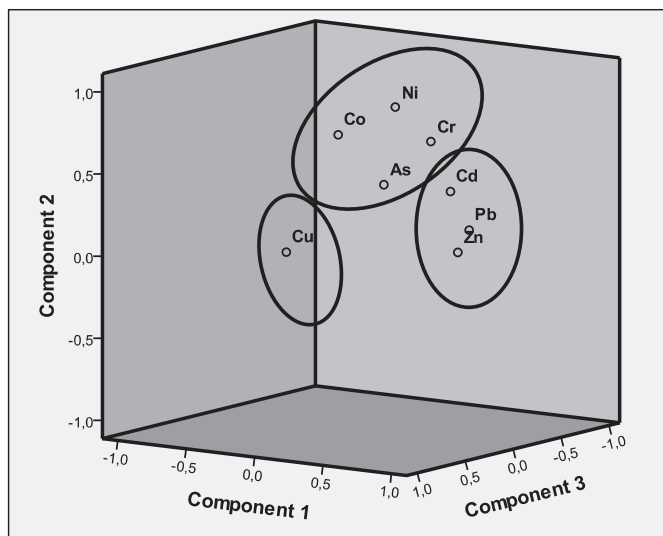


Fig. 7. The metals in the three-dimensional space of the three principal components, after rotation.

concentration can be detected in vegetable gardens and orchards than in flower gardens because of more intensive pesticide use. This is in line with the findings of Chen et al. (1997), who found also elevated Cu concentrations in orchard and vegetable soils of Hong Kong arising from the extensive use of these agrochemicals. Furthermore, Cu spatial distribution shows that Cu concentration is higher in samples distant from the road than close to it. Consequently, it can be established that pesticide use can predominantly be the cause of anthropogenic enrichment.

5. Conclusions

Various human activities (e.g. intensive cultivation: addition of organic matter such as compost, soil improvers, fertilizers; frequent infilling, mixing, construction, suburban sprawl, etc.) modify urban garden soils on the outskirts of Szeged, resulting in diverse physical and chemical soil properties. In addition to the soil characteristics, the metal concentration of urban garden soils is also modified owing to the direct and indirect impacts.

Despite the fact that urban garden soils have slight metal contamination, this study serves as a precedent on how the metal origin (geogenic and anthropogenic) can be differentiated and the anthropogenic source can be identified by means of combining different methods: enrichment factors (TEF, EFP), statistical analysis (Principal Component Analysis, ANOVA, *t*-test) and spatial distribution of the metals visualized by GIS.

According to the statistical coincidence of EFs confirmed by the *t*-test, Cu, Zn and Pb were considerably enriched in the topsoils, whereas Ni, Co, Cr and As did not accumulate. PCA also revealed the geogenic origin of Ni, Co, Cr, and As and differentiated two groups of anthropogenic metals [Pb, Zn, Cd] [Cu], indicating their different sources. Although Cd exceeds the “B” limit value in some gardens due to point sources, it is slightly enriched in the topsoils. On the contrary, Pb and Zn never reached the “B” limit value and are significantly enriched in the topsoils. In addition to intensive pesticide and soil improver use, these metals may derive mainly from traffic emission. The anthropogenic origin of Cu is unambiguous since it exceeds the “B” limit value in many gardens on the one hand, and shows the highest enrichment in the study area on the other hand. By contrast with the above metals, Cu originates predominantly from copper pesticides rather than traffic, in addition

to which there is a significant difference in its concentration depending on garden type.

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